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14. ABSTRACT
We have been very productive during the entire funding period. We have reached all three goals listed in the project title well beyond the initial expectations and have published a number of high profile papers documenting these successes. We have given numerous invited talks on the research supported by AFOSR. We have achieved direct stabilization of a femtosecond-laser-based optical frequency comb by a high-finesse passive optical cavity. We carried out detailed comparison of two distinct stabilization schemes and the result leads to new understanding of the optimum conditions for cavity stabilization as well as on approaches to overcome limitations on the ability to transfer the frequency stability of the cavity to the microwave domain. The stability of the frequency comb is explored in both the optical and the radio frequency domain. With an independent, stable CW laser, we have verified that the line width and stability of the wide-bandwidth optical comb components, respectively, reach below 300 Hz and 5 x 10-14 at 1 s averaging time, both limited by the reference CW laser.

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Final performance report to AFOSR on Project FA9550-03-1-0415: "Control and storage of femtosecond pulses via passive optical cavities – ultrastable ultrafast lasers, gain-less passive amplifiers, and ultrasensitive wide-bandwidth laser spectroscopy"
Principal Investigator: Jun Ye, University of Colorado, Boulder, Colorado

We have been very productive during the entire funding period. We have reached all three goals listed in the project title well beyond the initial expectations and have published a number of high profile papers documenting these successes. We have given numerous invited talks on the research supported by AFOSR.

First, we have achieved direct stabilization of a femtosecond-laser-based optical frequency comb by a high-finesse passive optical cavity. We carried out detailed comparison of two distinct stabilization schemes and the result leads to new understanding of the optimum conditions for cavity stabilization as well as on approaches to overcome limitations on the ability to transfer the frequency stability of the cavity to the microwave domain. The stability of the frequency comb is explored in both the optical and the radio frequency domain. With an independent, stable CW laser, we have verified that the linewidth and stability of the wide-bandwidth optical comb components, respectively, reach below 300 Hz and 5×10^{-14} at 1 s averaging time, both limited by the reference CW laser. Such a performance represents the state-of-art in frequency/phase stabilization of a femtosecond laser and fully confirms the expectation that a highly stable, passive optical cavity can directly stabilize the repetition frequency and carrier-envelope phase of ultrashort pulses to a level that rivals that achieved by cw lasers. These results are reported in a Rapid Communication paper in Physical Review A.

A second exciting development is that we have demonstrated a general technique for enhancement of femtosecond pulses from a pulse train through their coherent buildup inside a high-finesse cavity. Periodic extraction of the intracavity pulse via a fast switch provides a net energy gain of 42 to more than 70 times for 38 to 58 fs pulse durations. Starting with an actively stabilized, but otherwise standard mode-locked laser system, over 200 nJ pulses are demonstrated. Active control and stabilization of a fs laser's two degree's of freedom enables buildup and storage of ultrashort pulses in high-finesse optical cavities. The stored intracavity field is the result of the coherent addition of pulses accumulated over the lifetime of the cavity. A fast acousto-optic modulator (AOM) is used to switch out the enhanced pulse. Greater single-pulse amplifications are achieved using higher-finesse cavities (longer cavity lifetimes) at the expense of a reduction in the pulse repetition rate. Likewise, higher repetition rates can be maintained with more modest pulse amplifications. This result was published in Optics Letters.

There is a range of applications that can benefit from simple, cost effective systems capable of producing pulse energies in the μJ to mJ range. Many applications would also benefit from higher repetition rates for enhanced signal acquisition capabilities while others simply do not require the excessive pulse energies produced with conventional amplifier systems at the cost of added complexity. Our approach represents a flexible and general technique for enhancement of fs pulse energies by more than two orders of magnitude, applicable to ultrashort pulse trains in any spectral region where appropriate mirrors can be produced. This approach also enables generation of 100's of nJ pulse energies using common fs laser designs or could be used to compliment recent high-energy fs oscillators to potentially produce pulse energies >10 μJ. Besides this more conventional aspect of pulse amplification, the passive fs buildup cavity allows a new, exciting opportunity to explore nonlinear optical interactions in an intracavity environment at the laser's original high repetition rate. Nonlinear dynamics inside a femtosecond buildup cavity were reported in an Optics Express article.

Thirdly, we have also developed a precision measurement protocol to characterize mirror dispersion properties. The measurement accuracy has improved by more than ten fold over the previous state-of-the-art white light interferometry technique. We have provided these precisely measured results to the mirror coating manufacturer and allowed the company to significantly improve its capability in producing laser bandwidth, low loss and low dispersion mirrors. These results were summarized in an Optics Express article.

Two important research directions have since been since opened based on the aforementioned work. As it becomes increasingly challenging to maintain phase coherence beyond multiples of seconds, it is natural that we look beyond the visible domain and consider speeding up the "wheel of precision measurement" to the next level of carrier frequency. We have pursued two related experimental directions to address this vision. One is the generation of phase coherent frequency combs in the VUV (50 - 200 nm) spectral domain. In parallel, we are pursuing direct frequency comb spectroscopy to ready ourselves for quantum optics and precision spectroscopy once phase coherent sources become available in VUV. Both experiments benefit from the use of femtosecond enhancement cavities. These are passive optical cavities with high finesse and low dispersion over a large spectral bandwidth such that incident femtosecond pulse trains can be efficiently coupled inside. Pulse energies can be enhanced by three orders of magnitude to >10 μJ while the original pulse repetition frequency is maintained. This capability permits phase coherent high harmonic generation process to take place at enhanced average efficiency. We have successfully produced high harmonic generations at high repetition frequency (100 MHz) and the ground-break results were published as a cover article in the Physical Review Letters.

In addition to the power enhancement aspect, femtosecond cavities effectively increase the interaction length between matter and light, allowing direct frequency comb spectroscopy to acquire linear or nonlinear atomic and molecular signals with dramatically increased sensitivity. Hundreds of thousands of optical comb components, each coupled into a specific cavity mode, collectively provide sensitive intracavity absorption information simultaneously across 100 nm bandwidth in the visible and near IR spectral region. By placing various atomic and molecular species inside the cavity, we have demonstrated real-time, quantitative measurements of the trace presence, transition strengths and linewidths, and population redistributions due to collisions and temperature changes. This novel capability to sensitively and quantitatively monitor multi-species molecular spectra over a large optical bandwidth in real-time provides a new spectroscopic paradigm for studying molecular vibrational dynamics, chemical reactions, and trace analysis. This pioneering effort has resulted in a high profile paper published in the journal Science and it has attracted a great deal of attention from both industrial and academic researchers.

In summary, this project has been spectacularly successful. It has opened many new exciting applications that are ready for us to explore. We are proud to declare that we have opened a new subfield in ultrafast science and technology. We greatly appreciate the support from AFOSR.